# A Volumetric Memory Device based on Photo-Chromatic Compounds

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*Abstract* - **Three-dimensional (3D) optical memory is a revolutionary technology that has the benefits of lower cost (tens of dollars/Gbit), order of magnitude smaller size and mass, and low risk. We report on investigation and characterization of a three-dimensional (3D) volumetric optical memory device based on a new class of light-absorbing (photo-chromic) compounds that, when pulsed with lasers, absorb photons two at a time and can trigger chemical and physical changes (such as fluorescence) with micrometer-sized resolution in three dimensions.**

## I. INTRODUCTION

Commercial applications stored one exabyte ($10^{18}$ Bytes) of information last year, and this number is growing to more than two exabytes by the year 2002. Approximately 10 % of the information will be stored on magnetic disk drives, with the remainder on tapes and optical disks. This increasing capacity demand has thus far been met through steady increases in areal density of magnetic and optical recording media, where data are stored on a plane surface. While the limits of magnetic recording are still being debated - recently 100 Gbit/in$^2$ has been demonstrated - the limits of conventional optical storage are well understood. Current optical storage technology is working close to the diffraction limit (5 Gbit/in$^2$ ). Future increases in density are possible by taking advantage of shorter wavelength lasers, higher lens numerical aperture, or by employing near-field techniques. Conventional optical data storage capacities have also been increased by creating 2-layer media and bonding two such media back to back.

The three-dimensional approach to increasing the effective storage capacity is quite unique for optical memory technologies. Three-dimensional storage is envisioned as a cubic storage element with bit spacing having dimensions of the writing/reading laser wavelength. Instead of recording only on a plane, bits are stored throughout the volume of the material. With a wavelength of 650 μm, storage of one terabit per cubic centimeter is possible.

We are investigating and characterizing a three-dimensional (3D) and volumetric optical memory device based on a new class of light-absorbing (photo-chromic) compounds that, when pulsed with lasers, absorb photons two at a time and can trigger chemical and physical changes (such as fluorescence) with micrometer-sized resolution in three dimensions.

Our activities aim at enabling the Earth Science Enterprise (ESE) to integrate large global data sets involving space based observation systems. With many space and ground-based sensors, ESE must acquire, process, and deliver huge volumes of data. These remote sensing and related data will help solve problems such as global change, environmental monitoring, agricultural inventory, etc. The amount of data to be collected and processed per satellite runs into terabytes.

Development of adaptive, high capacity, high data rate optical storage for space vehicles can set an infusion path by establishing a realistic on-board storage platform for such data organization technologies as feature extraction and data prioritization for transmission. Impact of establishing a reliable ultra high capacity storage can also significantly influence development of hierarchical data segments, and reduction in data volume for downlink. Terabyte capacity on-board optical storage

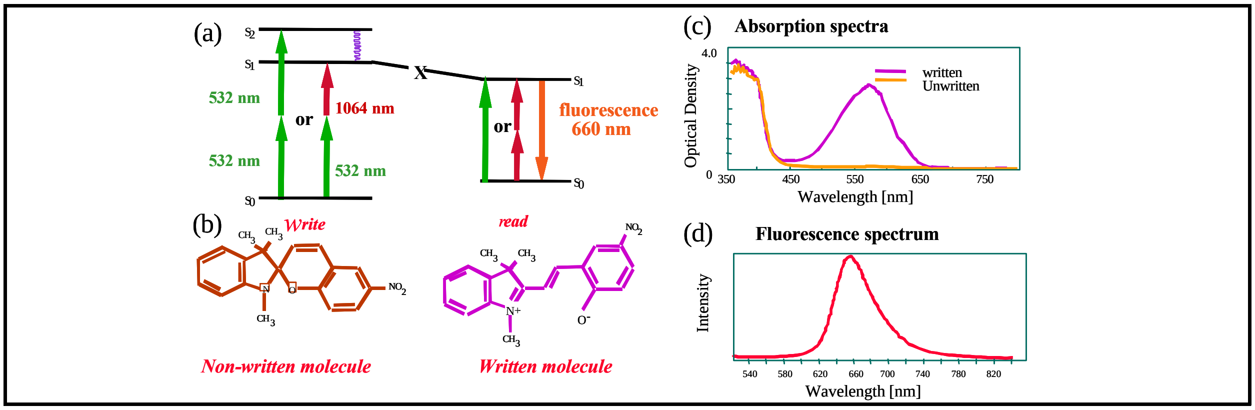


**Figure 1**. Example of a photochromic/polymer memory media: SP in PMMA; (a) energy state transition diagram; (b) written and unwritten molecular forms;(c) written and unwritten absorption spectra; (d) written form fluorescence spectra.

is an ideal platform on which generation of data products for direct distribution to users can happen.

The following sections provide a brief introduction to his technology and a snapshot of our current research progress. Section 2 discusses details of the photochromic process and the format of our test samples. Section 3 describes characterization of the linear and nonlinear optical parameters. Section 4 describes simulation of the optical readout process. Section 5 shows our first results from the readout test stand, and Section 6 summarizes our conclusions.

## II. THE PHOTOCHROMIC PROCESS AND DISK FORMAT

With tightly focused lasers, the photochromic process can be initiated and controlled within micrometer-size spaces. As shown in Figure 1, for instance, in the approach under development at Call/Recall, Inc.,[1] and the University of Arizona, a spot is written in the volume of a molded photochromic dispersed organic polymer only at points of sufficiently high intensity. This can be achieved by sharply focusing a single beam, or at the temporal and spatial intersection of two beams with sufficient photon energies, one carrying information, and the other specifying location. The recorded bits are read by fluorescence when written molecules within the written spot volume are excited by absorption of single green or red photons.

2-photon absorption makes it possible to write selectively a single bit anywhere within a three-dimensional volume by simply intersecting two optical beams, with the appropriate energies. When two photons of $\lambda_1$ and $\lambda_2$ wavelengths interact simultaneously, they are absorbed, resulting in a bond dissociation. Thus, the molecular structure is changed into a new, 'written', molecule with a different absorption and emission spectrum. To "read" the information written within the volume, the approach exploits the fact that the written form absorbs at longer wavelengths than the unwritten form. As shown on the right side of Fig. 1, the excitation of the written molecules is followed by a fluorescence at ~660 nm, which returns the molecule to its ground state. The presence or absence of this fluorescence is detected and classified as a logical '0' or '1' for the stored bit. Since the decay lifetime is ~5 nanoseconds and the concentration of molecules is high, it is possible to excite the written molecules many times in a single read cycle and increase the total light collected at the detector.

The advantages of the 2-photon absorption process are based upon its ability to selectively excite molecules inside a volume without populating molecules on the surface of the device. This may be achieved because the wavelength of each beam is longer and has less energy than the energy gap between the ground state and first allowed electronic level. Therefore, it propagates through the medium without being absorbed by a one-photon process. However, if two beams are used for excitation and the energy sum of the two interacting photons is equal to or larger than the energy between the ground and first excited state ($S_0 \rightarrow S_1$), absorption will occur. There is no real level at the wavelength of either beam, therefore neither beam can be absorbed alone by a one-photon mechanism. When the two photons collide within the volume, absorption occurs only within the volume of interaction and the size of the written bit is defined by the width of the two beams. The transition probability of a 2-photon absorption process partly depends upon the product of the two writing beam intensities. Thus, lasers emitting high intensity light in short pulses, i.e. picosecond and sub-picosecond pulses must be used. The recording material is dispersed in a polymer host which can then be shaped to produce disks with integrated structures for alignment and mounting. This project uses 1"x 0.25" PMMA disks with homogeneously dispersed storage materials. Polymerization molding, compression molding and polishing have been utilized to produce the desirable optical quality polymer for 3D optical memory disks.

A representation of the disk we use is shown in Fig. 2, where other technologies are also shown for comparison. Our one-inch test samples, if used with 500 layers and 1 Gbyte per layer, can produce a disk containing 500 Gbytes. With parallel readout beams, a high data rate can also be achieved.

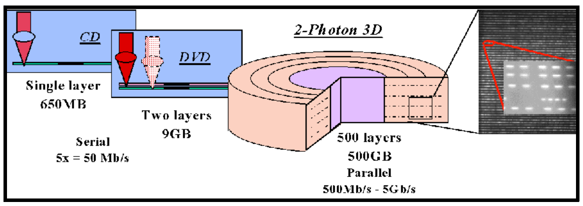


**Figure 2**. Technology for 2-photon recording is capable of producing volumetric media with very high capacities. For example, if our one-inch test disk is used with 500 data layers and 1 GByte per layer, the total capacity is 500 Gbytes. With parallel readout beams, a high data rate can also be achieved.

## III. OPTICAL MATERIAL MEASUREMENTS

The recording material is an essential element in the optical data storage system. Both the behavior of materials currently being used and the characterization of promising new materials are measured in this study. Optical characteristics of the materials are studied in detail.

The basic relationship governing the absorption of energy in 2-photon media is given by Beer's Law in the form

$$\frac{dI}{dz} = -\alpha I - \beta I^2 \, , \qquad \textbf{(1)}$$

where I is optical irradiance in $W/m^2$, α is the linear absorption coefficient and β is the nonlinear absorption coefficient. The value of α defines the amount of light absorbed in the material at low light levels. For our materials, α is small compared to β.

It is the parameter β that influences the ability of our laser to write data deep inside the medium without being affected by the data layers the beam passes through on it's way to the focus point. This property is illustrated in Fig. 3, which shows that the change in media properties during the single-beam writing process only occurs at the focus.

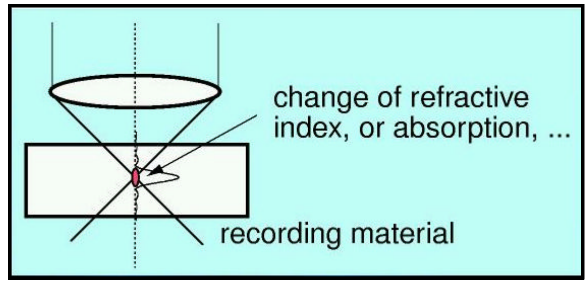


**Figure 3**. The change in material properties during the writing process occur only near the focus of the laser beam, where the irradiance is high enough to affect 2-photon absorption.

It is important for our research to understand how α and β are affected by environmental factors that may be found in space, such as temperature fluctuations and exposure to ionizing radiation. The reason for such variations is that β is dependent upon the complex susceptibility coefficient of the given material, as given by Eq. (2) in MKS units. Eq. (2) is easily derived from the general form of the Kramers-Krönig relations [2]..

$$\beta = \frac{3\omega}{2 n_0^2 \varepsilon_0 c} \mathrm{Im}\{\chi(3)\} \qquad \textbf{(2)}$$

The value of α is easily obtained from simple absorption measurements, but experimental values of β are more difficult to measure. The most usual method is the Z-Scan technique [3], which uses a single laser beam and a curve fit to measure both α and β. A variation on the Z-scan also yields the nonlinear refractive index of the material of interest $n_2$ quite accurately. This measurement is quite important, for nonlinear interactions of light with matter, much like their isotropic counterparts, never occur isolated from one another. A potentially good candidate material with high β must also have its nonlinear refraction properties known at the wavelength of interest, so effects such as optical limiting and self-focusing inside the material can be taken into account when designing any data storage scheme to utilize such media.

A characterization test stand facility is being constructed for the purpose of measuring α and β. Material-oriented studies are concentrating not only on searching for materials with high β, but also in searching for best matches between materials and feasible laser sources to be used with a given medium. Material cost, reliability and ruggedness under field conditions must also be a factor in those selections.

## IV. SIMULATION OF OPTICAL READOUT

Simulations for the 2-photon data storage read out system are accomplished with OPTISCAN, which is a beam-propagation engine and analysis tool developed by our research group.[4] Our first-order model assumes scalar, polarization-independent propagation in the medium and ignores media scattering effects. Despite the initial simplicity of our current model, the calculations provide good insight into such systems. Parameters already investigated include signal contrast and crosstalk between different data layers and in different materials.

Two simulations are complete that examine both confocal and non-confocal read out systems. The first simulation scans through a single-layer with deep marks, as shown in Fig. 4. A simple beam propagation technique was used to calculate the irradiating and collection energy from the mark pattern. The first simulation predicts high contrast for both confocal and non-confocal read out systems.

The second simulation extends the study to include three layers, where interlayer crosstalk is investigated. The optical geometry of the multi-layer mark pattern is shown in Fig. 5. In Case 1, a high-frequency data layer is surrounded by two low-frequency data layers. In Case 2, an isolated high-frequency layer is scanned. The difference between the readout signals for Case 1 and Case 2 is an indication of the inter-layer crosstalk. The confocal system is found to have better crosstalk rejection.

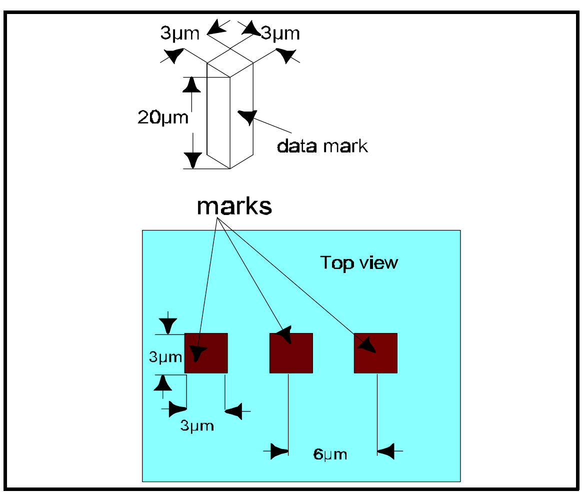


**Figure 4**. The first simulation study uses 3µm square and 20 µm deep data marks.

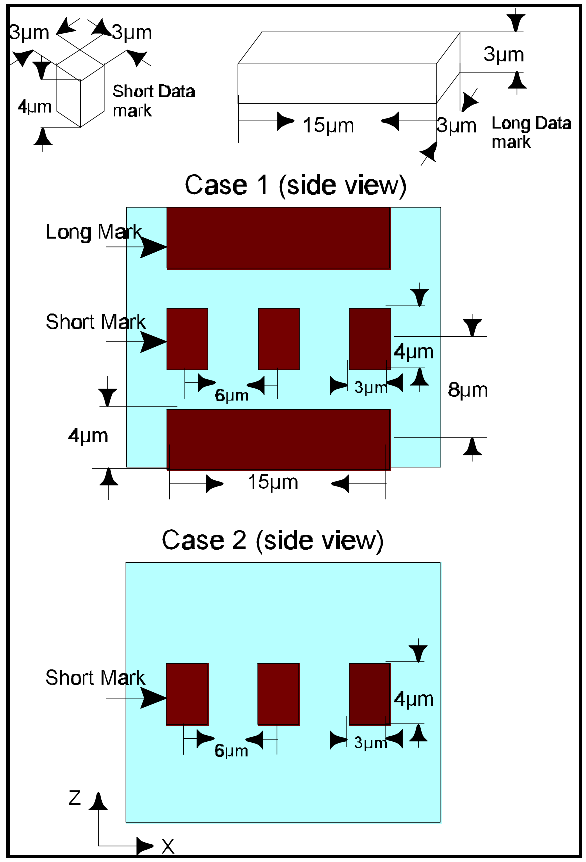


**Figure 5**. In the second simulation study, inter-layer crosstalk is studied by performing scans with a layer surrounded by two low-frequency layers (Case 1) and an isolated layer. The difference in readout signal between th e two scans is due to the crosstalk from the low-frequency marks.

## V. OPTICAL READOUT TEST STAND

A dynamic test stand is constructed in order to develop readout channels and associated electronics, as shown in Fig. 6. The test stand is based on a semi-kinematic rail system developed by our group.[5] Opto-mechanically, it consists of 4 inch x 4 inch stages, V-groove stages aligned along a fixed alignment bar, whose position is well-known. The diode laser at wavelength 638 nm and two alignment mirrors are placed in the first two 4 inch x 4 inch stages. The laser beam is then reflected 90 degrees by a dichroic beamplate which is placed in the third 4 inch x 4 inch stage along with a high-pass (650 nm cutoff wavelength) filter. This laser beam is then delivered through a relay telescope, also positioned with the help of the semi-kinematic V-groove system to a Geltech lens mounted on a Sony actuator. The beam is then focused into a disk provided by Call/Recall, Inc.

The marks inside the disk absorb the energy

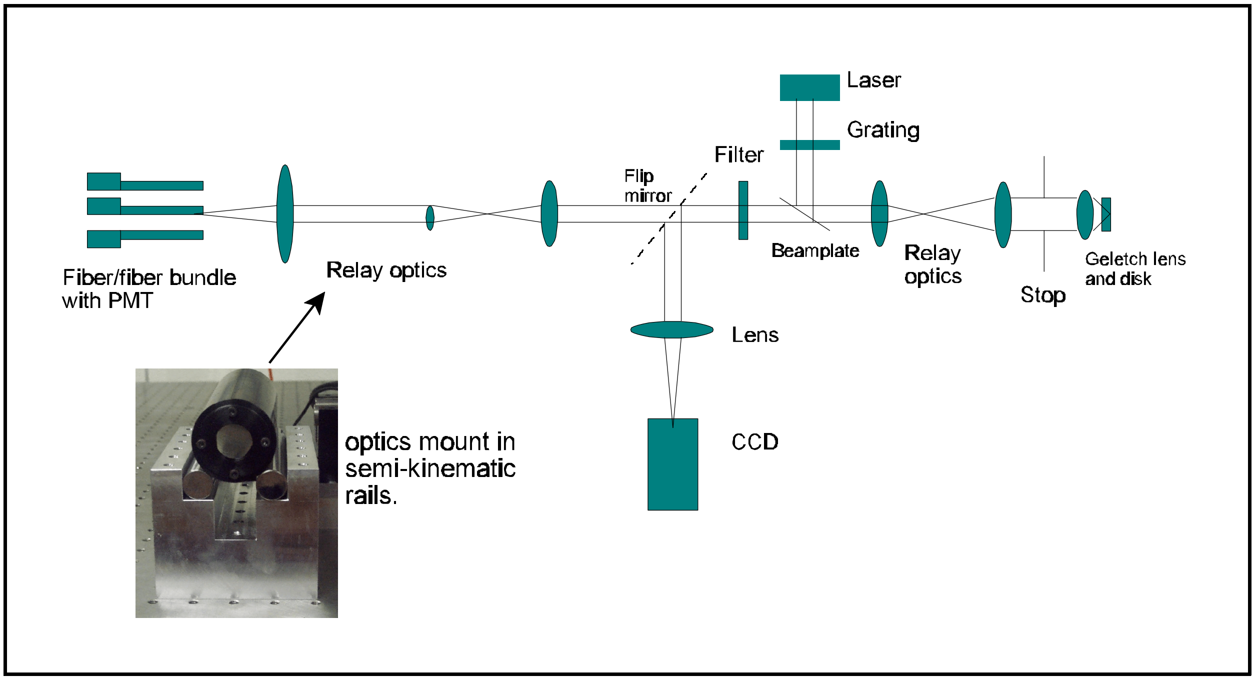


**Figure 6**. Layout of the 2-photon readout test stand.

and emit the fluorescent light whose spectrum ranges from 650nm to 700nm. The dichroic beam plate combined with a high band filter are used to separate the reflected laser beam and the fluorescent beam. A flipper mirror is used to direct the fluorescent beam to two different paths. In the first path, the fluorescent beam is focused into a black and white CCD camera. In the second path, a lens is used to focus the light into a bifurcated fiber bundle to collect the fluorescent signals and transmit the signals to PMTs. By measuring the SNR, we can optimize the mark size and the distance between each layer. The test stand is also used to characterize the signals changes observed when the material is exposed to space environments. Fig. 7 shows an image of the marks inside the disk as seen by the CCD camera mentioned above.

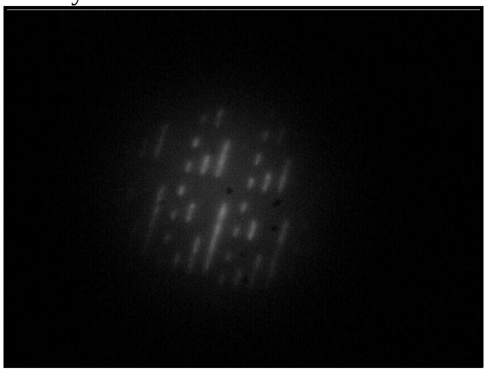

**Figure 7**. An image of 2-photon marks on a test disk as imaged by the CCD camera.

## VI. SUMMARY AND CONCLUSIONS

In summary, 2-photon data storage offers a tremendous improvement in capacity and other factors important for space applications. Our research is highly leveraged from other efforts in this area, and we are developing the technology through our understanding of media characteristics, simulations, and readout experiments.

## ACKNOWLEDGEMENT

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